CLAIM + DETAILED DESCRIPTION

[Claim(s)]

[Claim 1] The manufacture method of the polymer electrolyte fuel cell characterized by exfoliating said base material from said ion-exchange membrane after applying the liquid containing ion-exchange resin and a catalyst to the ion-exchange membrane fixed to the base material and drying.

[Claim 2] The manufacture method of a polymer electrolyte fuel cell according to claim 1 that the exfoliation adhesion strength of the ion-exchange membrane fixed to said base material and said base material is 0.005 - 0.1 kN/m under the conditions of **** speed 50 cm/min in the 180-degree exfoliation examination specified to JIS-K6829.

[Claim 3] Each ion-exchange resin contained in the resin which constitutes an ion-exchange membrane, and said liquid is the manufacture method of the solid polymer electrolyte type fuel cell according to claim 1 or 2 which consists of a perfluorocarbon polymer which has an ion exchange group.

[Claim 4] Said liquid is the manufacture method of the polymer electrolyte fuel cell according to claim 1, 2, or 3 which is applied to an ion-exchange membrane in normal temperature, and is dried at the temperature of the range from normal temperature to 180 degrees C.

[Claim 5] The manufacture method of the polymer electrolyte fuel cell according to claim 1, 2, 3, or 4 which applies the liquid which takes out the field where said liquid of said ion-exchange membrane is not applied to a table, fixes to another base material, and contains ion-exchange resin and a catalyst in said field after exfoliating said base material from said ion-exchange membrane, and is dried.

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the manufacture method of a polymer electrolyte fuel cell.

[0002]

[Description of the Prior Art] A reaction product is only water theoretically and hydrogen and an oxygen fuel cell attract attention as a power generation system which does not almost have a bad influence to earth environment. Power density is becoming high by rapid progress of research in recent years, and utilization is expected very much especially from the polymer electrolyte fuel cell.

[0003] In a polymer electrolyte fuel cell, an electrode is close with the ion-exchange membrane which is a solid polymer electrolyte, arranges and has the layer (catalyst bed) including a catalyst, and has a gas diffusion layer for supplying gas to the outside of a catalyst bed efficiently at a catalyst bed if needed. On these Descriptions, an ion-exchange membrane and a catalyst bed are close, a gas diffusion layer is arranged, without including, and what is joined on appearance is called an electrode and film zygote.

[0004] After forming a catalyst bed on the base material which serves as gas diffusion layers which form a catalyst bed directly on an ion-exchange membrane, such as a method and a carbon paper, as the manufacture method of of the conventional electrode and film zygote, the method of joining this to an ion-exchange membrane, the method of forming an electrode on a plate and transferring this to an ion-exchange membrane, etc. are. By these methods, in order to form a catalyst bed, the liquid (henceforth catalyst ink) which distributed the catalyst is used. A **** agent and a thickener are usually contained in catalyst ink if needed [the catalyst powder, the ion-exchange resin, ******, and if needed] which supported platinum group metal to activated carbon etc., and these are distributing or dissolving in the solvent.

[0005] By the method of applying direct catalyst ink to an ion-exchange membrane, it usually applies by the existing methods, such as DAIKOTA and screen-stencil. In order to make a catalyst bed adhere to an ion-exchange membrane, it is effective in catalyst ink to make the solvent ingredient which dissolves an ion-exchange membrane ingredient contain. However, catalyst ink composition cannot adjust strictly, but when the film lytic reaction by a solvent is too strong, or in using the ion-exchange membrane which is not reinforced in three dimensions with a porous object etc., by applying catalyst ink, an ion-exchange membrane swells and wrinkles occur. Therefore, there is a problem from which an electrode and a film zygote with a smooth bonded surface are not obtained.

[0006] So, in this invention, whether it uses a non-reinforcing film with bad dimensional stability for an ion-exchange membrane or uses the catalyst ink containing the strong solvent of the operation which dissolves and swells an ion-exchange membrane, wrinkles do not occur on a film but an electrode and a film zygote with a smooth bonded surface of a film and an electrode are obtained. Therefore, it aims at offering a high-output polymer electrolyte fuel cell.

[0007]
[Means for Solving the Problem] After this invention applies the liquid containing ion-exchange resin and a catalyst to the ion-exchange membrane fixed to the base material and dries, it offers the manufacture method of the polymer electrolyte fuel cell characterized by exfoliating said base material from said ion-exchange membrane.
[0008]
[Embodiment of the Invention] In this invention, generating of membranous wrinkles is controlled by applying the catalyst ink containing ion-exchange resin and a catalyst to the ion-exchange membrane fixed to the base material. Therefore, the ion-exchange membrane fully needs to be fixed to the base material. That is, since the film surface will dissolve in the solvent ingredient of catalyst ink if catalyst ink is applied to the ion-exchange membrane fixed to the base material, it is going to change, and a film tends to swell, and tends to exfoliate from a base material, and wrinkles generate it on a film. Therefore, the power in which a base material fixes a film needs to be stronger than this power in which it exfoliates.
[0009] It is desirable to fabricate a film on a base material, for example at the time of membrane formation of an ion-exchange membrane, and to use a film as it is as an ion-exchange membrane made to fix to a base material, without exfoliating from a base material. The ion-exchange membrane which is obtained by the cast method which specifically casts the solution which dissolved ion-exchange resin in the solvent, for example on a direct base material, and is dried and which was fabricated on the base material can be used the whole base material. Moreover, even if it makes a film become wet and lays on a base material, depending on the membranous quality of the material, it is fixable on a base material.
[0010] Moreover, when a film functions as an electrolyte of a fuel cell, as long as it does not have a bad influence, you may fix a film to a base material using adhesives. Moreover, you may fix a film to a base material on a tape etc. As the above-mentioned film and a tape which exfoliates easily, the Sumitomo 3M Post-it cover rise tape (brand name) etc. is desirable, for example, and it is desirable to be fixed on a film and the tape which exfoliates easily, where a film is pulled.
[0011] It pastes up moderately with a film and the base material which fixes an ion-exchange membrane has a film and the desirable thing which can exfoliate easily. Moreover, it has the chemical resistance for not carrying out an interaction to a film chemically, but keeping a stable state chemical, and the moderate thickness which is easy to handle, surface coarseness is small, and it is desirable to have the rigidity which can bear the stress which a film tends to transform with adhesives. Moreover, since it may be required when heating dries catalyst ink, and it may be required also when a film is fixed to a base material, as for a base material, it is desirable to have heat resistance.
[0012] Although the form of a base material has the ease of carrying out of an application of

catalyst ink to a desirable plate, since what is necessary is just to have the function which fixes a film until it dries after catalyst ink's applying, it may be a curved surface. For example, a film can be fixed to a roller-like base material and catalyst ink can also be applied.

[0013] When the 180-degree exfoliation examination specified to JIS-K6829 is applied correspondingly and **** speed is made into the conditions of 50 cm/min, as for the exfoliation adhesion strength of the ion-exchange membrane and base material which are being fixed, it is desirable that it is especially 0.01 - 0.06 kN/m 0.005 to 0.1 kN/m. By less than 0.005 kN/m, an ion-exchange membrane is not fully fixed to a base material for adhesion strength, but on the other hand, adhesive strength is too strong and exfoliation of an ion-exchange membrane and a base material is difficult at more than 0.1 kN/m.

[0014] As for especially the thickness of a base material, 50-250 micrometers is desirable 10-350 micrometers from a point of handling nature. Moreover, since an ion-exchange membrane is fixed to a base material, even if it has a projection, it is desirable [coarseness / as for the surface coarseness of a base material, it is desirable that the variation in thickness is less than **5%, and] that the height is 0.03 micrometer or less.

[0015] It is desirable to have tolerance to organic solvents, such as sulfuric acid, and ethanol, acetone, as chemical resistance of a base material 10%. Moreover, it is desirable that it has the heat resistance of 150 degrees C or more, and can hold for several minutes also in 200 degrees C as heat resistance of a base material. It is for heating according to **** of a solvent in the dryness process for the solvent removal after a catalyst ink application.

[0016] moreover, [point / that excelling in rigidity is desirable as for a base material, and rigidity is the function of an elastic modulus and thickness] It is desirable that it is 400-6000Ns/mm2 when the **** elastic modulus specified to JIS-K7127 examines by test-rate 200 mm/min in a specimen 15mm in width and 200mm in length.

[0017] If an example desirable as a base material is given, a polyethylene terephthalate (henceforth PET) film, a polyolefin film, tetrafluoro ethylene / ethylene copolymer (henceforth ETFE) film, a polyimide film, etc. will be mentioned. Moreover, the glass board made from Pyrex (brand name) of an inorganic substance is also desirable.

[0018] Especially if it is not the temperature to which the solvent in catalyst ink volatilizes in an instant, it will not be limited, but the ambient temperature at the time of applying catalyst ink to a film has desirable normal temperature, when workability and equipment are taken into consideration. Moreover, although it is dependent on **** of the solvent in catalyst ink, it is necessary to make the conditions of the dryness for removing solvent removal after a catalyst ink application into a temperature lower than a heat-resistant temperature of the ion-exchange resin contained in an ion-exchange membrane and a catalyst bed. It is desirable to specifically consider it as the temperature of the range of normal temperature -180 degree C, and it is desirable to consider it especially as 150 degrees C or less.

[0019] Moreover, as for especially drying time, 5 minutes - 3 hours are desirable 10 seconds or more. When drying time is shorter than 10 seconds, a solvent may remain and a base material and a film are made to exfoliate, a possibility that wrinkles may occur is on a film under the influence of a residual solvent. On the other hand, when drying time is longer than 3 hours, the ion-exchange resin which manufacturing efficiency worsens and is contained in an ion-exchange membrane and a catalyst bed receives a bad influence chemically, or there is a possibility of changing structurally and forming high resistance.

[0020] Although it changes with application equipment to be used, when using a screen printer, for example, as for the desirable characteristic of the catalyst ink in this invention, it is desirable that viscosity is thousands cP(s). Especially as a solvent of catalyst ink, it is not limited, for example, water, alcohols, ether, a fluorine-containing solvent, etc. can be used.

[0021] In this invention, after applying a direct catalyst ink solvent to the ion-exchange membrane fixed to the base material, drying and forming a catalyst bed (henceforth the 1st catalyst bed), a base material is made to exfoliate from an ion-exchange membrane, and the electrode and film zygote by which the catalyst bed was formed in membranous one side are obtained. Subsequently, another catalyst bed (henceforth the 2nd catalyst bed) is formed or joined to the field which was being fixed to the base material of an ion-exchange membrane.

[0022] As a method of forming or joining the 2nd catalyst bed, various methods, such as the method of applying direct catalyst ink to an ion-exchange membrane and the method of forming a catalyst bed in one side of a gas diffusion layer beforehand, and joining a film to a catalyst bed by the pasting-up method (referring to JP,7-220741,A), the hot pressing method, etc., are employable. However, generally, as for the 1st catalyst bed, it is desirable to have porous structure so that it may be thinly formed on a film and may be easy to diffuse gas, but when the 2nd catalyst bed is formed by the hot pressing method, the porous structure of the 1st catalyst bed will be crushed.

[0023] Moreover, when forming a catalyst bed on a gas diffusion layer, many amounts of catalysts are usually needed rather than forming a catalyst bed on a film. In order to use a porous body in a gas diffusion layer generally, when applying catalyst ink to the surface, catalyst ink permeates into a gas diffusion layer to some extent. Therefore, since a catalyst bed is not formed in the gas diffusion layer surface if more catalyst ink is not applied, the amount of catalysts of the 2nd catalyst bed increases also by the pasting-up method or the hot pressing method, and it is not desirable. Therefore, the method of applying direct catalyst ink to an ion-exchange membrane in that a thin layer is obtained as how to form or join the 2nd catalyst bed, without applying pressure is desirable.

[0024] However, in the case of the method of applying catalyst ink to a direct film and forming the 2nd catalyst bed, since the base material which fixes a film does not exist, a film may dissolve and swell and change into the solvent contained in catalyst ink, and wrinkles may

occur. Therefore, in order for membranous both sides to obtain a film and an electrode zygote with a smooth bonded surface of a film and a catalyst bed, it is desirable to form the 2nd catalyst bed, where the film and electrode zygote in which only the 1st catalyst bed was formed are fixed.

[0025] If a film is a rectangle, for example, it is in the state which fixed the crossroads or neighborhood to the base material on the film, the tape which exfoliates easily, etc., and, specifically, there is the method of applying catalyst ink to the field in which the 1st membranous catalyst bed is not formed. As the above-mentioned film and a tape which exfoliates easily, the aforementioned Post-it cover rise tape (brand name) etc. is desirable, for example, and it is desirable to be fixed on a film and the tape which exfoliates easily, where a film is pulled. If the 2nd catalyst bed is formed by the application of catalyst ink and the solvent of catalyst ink is removed by this method, it will become the flat bonded surface which wrinkles hardly generate on a film and wrinkles do not generate on a film like the time of formation of the 1st catalyst bed.

[0026] In addition, even if the catalyst ink for forming the catalyst ink and the 2nd catalyst bed for forming the 1st catalyst bed is the same, it may differ. Moreover, the 1st catalyst bed is [fuel pole and oxidizer pole side] good also as a catalyst bed of which electrode. As a catalyst, platinum group metal, platinum alloys, etc., such as platinum, can be used. Although these catalysts may be used as they are as metal particulates, you may use the support catalyst which supported metal to carriers, such as activated carbon and carbon black.

[0027] [the ion-exchange resin contained in the resin which constitutes the ion-exchange membrane in this invention, and a catalyst bed] It is desirable that it is the perfluorocarbon polymer (the oxygen atom of ether bond nature shall also be included in addition with a perfluorocarbon polymer on these Descriptions in addition to the polymer which consists only of a carbon atom and a fluoride atom, for example) which has an ion exchange group. An ion exchange group may be a positive ion exchange group, or may be an anion exchange machine, and a sulfonic group, a HOSUHON acid radical, etc. are mentioned as a positive ion exchange group. moreover, even if the resin contained in a catalyst bed is the same as the resin which constitutes an ion-exchange membrane, it may differ.

[0028] Since the solvent contained in catalyst ink in this invention can join an ion-exchange membrane to a catalyst bed strongly if it contains the solvent which can dissolve an ion-exchange membrane, it is desirable. Moreover, ******, the thickener for adjusting viscosity, the dilution agent, etc. may be added by catalyst ink if needed.

[0029] [the 1st catalyst bed and 2nd catalyst bed / the film and electrode zygote formed on the ion-exchange membrane] It is desirable to arrange the gas diffusion layer which becomes the outside from a carbon paper, carbon crossing, etc., and it is desirable that arrange the separator which supplies gas to the outside further and functions also as a current collection

object, and a polymer electrolyte fuel cell is assembled. And the fuel gas containing hydrogen and the oxidant gas which contains oxygen on the oxidizer pole are supplied to a fuel pole, and it generates electricity by reacting.

[0030]

[Example] <Example 1 (EXAMPLE)> 100 micrometers in thickness (thickness variation: less than **5% and less than projection height:0.025micrometer) The ion-exchange membrane (brand name: FUREMION, the Asahi Glass Co., Ltd. make, 1.1mm Eq/g ion-exchange-capacity dry weight) with a thickness of 50 micrometers which becomes a heat-resistant temperature range-70 degree-C-150 degree C (260 degrees C of melting points) PET film from the perfluorocarbon polymer containing a sulfonic group was fixed. After the method of fixation immersed the film for 10 seconds into distilled water at room temperature, it stuck the thing in the state where the waterdrop adhering to the surface was wiped off lightly so that it might not become wrinkles on a PET film, and forced and fixed it with the hand-pushed roller lightly.

[0031] At this time, the exfoliation adhesion strength of an ion-exchange membrane and a PET film was 0.03 kN/m under the conditions which make **** speed 50 cm/min in the 180-degree exfoliation examination specified to JIS-K6829. In addition, the PET film used here was 3 kN (s)/mm2, when the **** elastic modulus specified to JIS-K7127 examined by test-rate 200 mm/min in a specimen 15mm in width, and 200mm in length.

[0032] Catalyst bed In the catalyst ink for forming, 1.1mm Eq/g ion-exchange-capacity dry weight, The ion-exchange resin which consists of a copolymer which consists of a polymerization unit based on CF2=CF2, and a polymerization unit based on CF2=CFOCF2CF (CF3) O(CF2)2SO3H, The liquid (a solvent is a mixed solvent of ethanol/water) which platinum support carbon (Pt/C=40/60 (mass ratio)) contains by 25/75 of mass ratios was used.

[0033] A catalyst bed can form the ion-exchange membrane fixed to the above-mentioned PET film only a 5.3cm angle. After masking using a screen printer, applied the above-mentioned catalyst ink so that the amount of platinum adhesion might be set to 0.4 mg/cm2, and it was made to dry at 80 degrees C after 1-hour dryness and among the air with room temperature among the air for 1 hour, and the 1st catalyst bed was formed. Subsequently, when the PET film was slowly exfoliated from the ion-exchange membrane, wrinkles were not looked at by the ion-exchange membrane, but the bonded surface of an ion-exchange membrane and a catalyst bed was smooth.

[0034] It laid in the state where it stretched so that the field where the 1st catalyst bed is not formed on the PET film with a thickness of 100 micrometers prepared separately in the sample by which the 1st catalyst bed was formed in one side of the above-mentioned ion-exchange membrane might be turned upwards and there might be no wrinkles. Subsequently, using the aforementioned Post-it cover rise tape (brand name), the neighborhood of the above-mentioned sample was stopped and the above-mentioned sample was fixed on the PET film.

[0035] Next, open and mask the space of a 5.3cm angle using a screen printer like the time of the 1st catalyst bed formation, and the amount of Pt adhesion should be set the above-mentioned catalyst ink to two in 0.4mg/cm. Applied, it was made to dry at 80 degrees C after 1-hour dryness and among the air with room temperature among the air for 1 hour, and the 2nd catalyst bed was formed. Then, although the tape on which the neighborhood of the sample was stopped was removed, wrinkles were hardly looked at by the ion-exchange membrane. Thus, the electrode and the film zygote by which the 1st catalyst bed and 2nd catalyst bed were formed in both sides of an ion-exchange membrane, respectively were produced.
[0036] Except having used it, without fixing the <Example 2 (comparative example)> ion-exchange membrane to a PET film, the 1st and 2nd catalyst beds were formed like Example 1, and the electrode and the film zygote were produced. In order to form the 1st catalyst bed, when catalyst ink was applied to a film, the wrinkles of about 0.5mm in thickness and the convex shape of a ring of 0.5mm of **** arose in tens of places in an instant at the ion-exchange membrane. After dryness, when taken out from the screen printer, kneading has already arisen into the portion in which the catalyst bed is not formed under the influence which the catalyst ink application portion contracted. Therefore, it was hard to form the 2nd catalyst bed, it was applied at the time of a catalyst ink application, and nonuniformity produced it. However, since the membranous neighborhood was fixed on the tape at the time of the 2nd catalyst bed formation, new wrinkles hardly increased.
[0037] [Evaluation] The electrode and film zygote obtained in Example 1 and Example 2 are used, and it is a gas diffusion layer to both the outside, The carbon crossing (the Japan Gore-Tex make, brand name:CARBEL-CL) formed in one side was included in the cell for measurement, after the carbon powder layer turned the carbon powder layer inside and had arranged it. Normal pressure, the cell temperature of 80 degrees C, and hydrogen/air were used for the operating condition of a cell, and the fuel capacity factor was made into 70% of hydrogen, and 40% of air. The terminal voltage of a cell and iR loss in current density 1.0 A/cm2 were measured. A result is shown in Table 1.
[0038]
[Table 1]

	セル電圧 (V)	iR損 (V)
例1	0.52	0.10
例2	0.45	0.13

[0039]
[Effect of the Invention] According to this invention, generating of the wrinkles of the film which originates in the operation in which the solvent in catalyst ink dissolves an ion-exchange membrane by performing the application process of the catalyst ink for forming a catalyst bed to the ion-exchange membrane which adhered on the base material is prevented, and smooth

electrode and film zygote are obtained by a simple method. The polymer electrolyte fuel cell which has this film and electrode zygote has a high output.

[Translation done.]